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On the theory of the diffusion cooling of neutrons in a finite solid moderator assembly[1]

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With 1 figure in the text

ABSTRACT

A general theory of the diffusion cooling of neutrons in a finite solid moderator assembly is given. In the present theory, starting from the transport equation in the diffusion approximation, the energy-dependent part of the solution is expressed in terms of generalized Laguerre polynomials of order unity and degree n, each term of which is weighted by a Maxwellian distribution function. In the approximation in which only the first two terms of the Laguerre expansion are retained, one obtains exactly the same expression for the decay constant of the fundamental mode as in the variational approach of Nelkin and of Singwi and Kothari. In the next higher approximation, one gets a correction which we call the "non-Maxwellian" correction to the diffusion cooling constant, which correction is nearly twenty to twenty-five per cent. The merit of the present theory is that it avoids the assumption of a neutron temperature which was the weak point of the variational approach. Expression for the correction to the diffusion cooling constant arising from terms of order B^6 has also been derived and it turns out that this correction has a sign opposite to that of the non-Maxwellian correction and in magnitude it is quite significant, depending on the value of the buckling, B^2 of the assembly. Non-diffusion correction to the decay constant has also been estimated and it is found that it is negligible compared to the multi-velocity correction. Expressions for the mean energy of neutrons inside and outside the assembly are derived. Finally the variation of the diffusion cooling constant with the temperature of the moderating assembly is studied in the two limiting cases: (i) $T/\theta \to \infty$ and (ii) $\theta/T \to \infty$, θ being the Debye temperature of the solid. Exact evaluation of this variation is shown graphically in the case of beryllium. Comparison of the theoretical values of the diffusion cooling constant with the experimental values for beryllium, beryllium oxide and graphite has been made. If the transport mean free path is assumed to be independent of energy, the agreement between theory and experiment is good in the case of graphite and beryllium, but bad in the case of beryllium oxide for which the experimental value seems to be rather high.

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Introduction

In an earlier paper, hereafter referred to as I, Singwi and Kothari (1958) had derived an expression (Eq. 8 of I) for the decay constant, λ of the fundamental spatial mode of the neutron flux, as a function of the geometrical buckling, B^2 of the moderating assembly. It is generally expressed in the form

$$\lambda = \sum_a v + \bar{D}_0 B^2 - C \bar{D}_0 B^4,$$

where the various symbols have their usual meaning. This expression is valid for times long compared to the slowing-down time of neutrons, which are injected into the assembly in the form of a pulse. The significance of the first two terms in the expression for λ is very well understood and there exists a fairly good agreement between theory and experiment as regards the value of the diffusion coefficient $\bar{D}_0$. This, however, is not the case with the last term in which the coefficient C is called the diffusion cooling constant. In a finite assembly there is a preferential leakage of fast neutrons; the rate of leakage depends on the size of the assembly and on the energy dependence of the diffusion coefficient. As a consequence of this, the neutrons inside the assembly cool below the moderator temperature. Thermal equilibrium will, however, be established as a result of the transfer of energy between the neutrons and the solid. The rate at which this equilibrium is established will depend on the efficiency of the mechanism of energy transfer. In fact the constant C is a measure of this efficiency. At low temperatures of the moderating assembly, the energy transfer is so slow that a thermal equilibrium may never be attained.

deSaussure and Silver (1958) have tabulated the experimental values of C as determined by different workers for beryllium at room temperature. It will be seen from their table that the values of C range from 0 to 3.05 ± 0.65 cm^2. Their own value is 1.1 ± 0.9 cm^2. The experimental position[1] is, therefore, very unsatisfactory. The theoretical value of C as calculated by Nelkin (1958) is apparently in good agreement with the experimental value of deSaussure and Silver. However, in a more realistic theory if one takes into account the variation of the transport mean free path with neutron energy this agreement is spoiled (paper I).

The earlier theories of Nelkin and of paper I were based on the assumption that the departure of the neutron energy spectrum from the equilibrium Maxwellian spectrum was small; and that one could associate a temperature T_n (if it exists!) with the neutrons. A trial function of the Maxwellian form for the neutron spectrum was taken with the neutron temperature T_n as a variational parameter. The Rayleigh-Ritz variational principle was used to determine the lowest eigenvalue of the decay constant. Nelkin examined the accuracy of the assumption of a neutron temperature by comparing his calculation with the exact results of Hurwitz *et al.* (1956) for the case of a heavy gaseous moderator.

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[1] For a general experimental discussion of the diffusion cooling constant see the review article (Progress in Nuclear Energy, Series 1, Physics and Mathematics, Vol. 2, Pergamon Press, London, 1958) by von Dardel and Sjöstrand.

His estimate was that the error in the diffusion cooling constant was of the order of twenty-five per cent. If the moderating assembly is small, there will be a large leakage of neutrons and consequently it becomes doubtful whether the association of a temperature to the neutrons inside the assembly has any physical meaning at all. The main purpose of this paper is to develop a more general theory in which any such assumption of a neutron temperature is completely avoided. Mention may be made here of a numerical calculation, based on a heavy gas model by Beckurts (1957), who found that considerable deviations from a Maxwell spectrum occur even for rather small bucklings.

The starting point of the theory is to expand the velocity distribution of the neutron flux in terms of generalized Laguerre polynomials of order unity and degree n, each one of which is weighted by a Maxwellian distribution function. Such an expansion has also been used recently by Kazarnovsky *et al.* (1958). The number of terms required in the expansion will obviously depend on the closeness of the actual spectrum to the Maxwellian spectrum. On retaining the first two terms of the Laguerre expansion, we get the same expression for C as obtained in I using the variational principle. The third term in the expansion gives a correction (to the order B^4) to the diffusion cooling constant which is estimated to be nearly twenty per cent. We would call this as the "non-Maxwellian" correction.

For an accurate experimental determination of C it is necessary that the moderating assembly be small, *i.e.* its buckling, B^2 be large. In such a case it is not possible to neglect terms of order B^6 in the expression for the decay constant. We have derived expressions for terms of order B^6 and B^8 in the approximation in which only the first two terms in the Laguerre expansion are retained. Expression for the term of order B^6 has also been obtained when the first three terms in the expansion are retained. It turns out that this correction has a sign which is opposite to that of the non-Maxwellian correction and it increases in magnitude at low temperatures. For the smallest beryllium assembly used by deSaussure and Silver, it is nearly fifty per cent of the non-Maxwellian correction to the diffusion cooling constant.

The magnitude of the "non-diffusion" correction to the cooling constant has also been estimated and it is found to be negligible compared to the non-Maxwellian correction.

Expressions for the mean energy per neutron in the flux inside and outside the assembly are given, which clearly indicate the effect of the variation of the transport mean free path with neutron energy. The inside energy, in general, differs from the outside energy; it is the latter which is measured experimentally.

The temperature dependence of the diffusion cooling conssant has been studied in the two limiting cases: (i) $T/\theta \to \infty$ and (ii) $T/\theta \to 0$, θ being the Debye temperature. In the latter case C varies as $(\theta/T)^6$, which indicates that for very low temperatures the diffusion cooling constant will be so large that thermal equilibrium may not be possible at all, whereas in the former C is constant. A graph showing the calculated variation of C with (θ/T) is also given. An experimental verification of this result would be interesting.

2. Mathematical formulation

The Boltzmann equation for the angular flux $f(\mathbf{r}, E, \mathbf{\Omega}, t)$ is

$$\frac{1}{v}\cdot\frac{\partial f(\mathbf{r}, E, \mathbf{\Omega}, t)}{\partial t} = -\mathbf{\Omega}\cdot \operatorname{grad} f(\mathbf{r}, E, \mathbf{\Omega}, t) - f(\mathbf{r}, E, \mathbf{\Omega}, t)\left[\Sigma_s(\mathbf{r}, E, \mathbf{\Omega}) + \Sigma_a(\mathbf{r}, E)\right]$$
$$+ S(\mathbf{r}, E, \mathbf{\Omega}) + \int dE'\, d\mathbf{\Omega}'\, f(\mathbf{r}, E', \mathbf{\Omega}, t)\, \Sigma_s(\mathbf{r}, E' \to E, \mathbf{\Omega}' \to \mathbf{\Omega}) \qquad (1)$$

(see Weinberg and Wigner (1958), p. 223), and where the symbols have their well-known meaning. v is the neutron velocity corresponding to energy E. Since our interest is in developing a theory for the pulsed neutron experiments, we shall take the source term $S(\mathbf{r}, E, \mathbf{\Omega})$ to be zero. In the P_1 (or the diffusion) approximation equation (1) takes the form

$$\frac{1}{v}\cdot\frac{\partial \Phi(\mathbf{r}, E, t)}{\partial t} - \frac{l_{tr}(E)}{3}\nabla^2 \phi(\mathbf{r}, E, t) + \Sigma_a(E)\,\phi(\mathbf{r}, E, t) + \phi(\mathbf{r}, E, t)\,\Sigma_s(E)$$
$$- \int_0^\infty \phi(\mathbf{r}, E', t)\,\Sigma_s(E' \to E)\, dE' = 0, \qquad (2)$$

where
$$\phi(\mathbf{r}, E, t) = \int d\mathbf{\Omega}\, f(\mathbf{r}, E, \mathbf{\Omega}, t).$$

The validity of Eq. (2), as is well known, is based on two approximations: (i) the neglect of the second and higher terms in the angular distribution of f and (ii) the neglect of the $\frac{\partial \mathbf{J}}{\partial t}$ term in the expression for $\mathbf{J}$, where $\mathbf{J}$ is the neutron current. We shall examine the influence of approximation (i) when we discuss the magnitude of the non-diffusion correction to the diffusion cooling constant, and as regards the second approximation, it is easy to show that it is valid so long as the scattering cross section is much greater than the absorption cross section.

By making a Laplace transform, the time variation can be separated from the transport Eq. (1), and the problem reduces to a time-independent equation, in which the time constant λ, or rather λ/v appears as an eigenvalue. This eigenvalue equation was solved in the diffusion approximation in paper I by using the variational method, assuming that the space and energy dependence of f can also be separated out.

Our starting equation is (2). Let us introduce a dimensionless variable $x = v^2/v_0^2$, where $v_0 = (2k_0 T/m)^{\frac{1}{2}}$, T is the moderator temperature, k_0 the Boltzmann's constant and m the neutron mass. Equation (2) then becomes

$$\frac{1}{\sqrt{x}}\frac{\partial \phi(\mathbf{r}, x, t)}{\partial t} - \frac{1}{3} l_{tr}(x)\,\nabla^2 \phi(\mathbf{r}, x, t) + \Sigma_a(x)\,\phi(\mathbf{r}, x, t) + \phi(\mathbf{r}, x, t)\int_0^\infty \Sigma_s(x \to x')\, dx'$$
$$- \int_0^\infty \phi(\mathbf{r}, x', t)\,\Sigma_s(x' \to x)\, dx' = 0, \qquad (3)$$

where in (3) we have put v_0 equal to unity.

Let ut assume that $\phi(\mathbf{r}, x, t)$ can be expressed as

$$\phi(\mathbf{r}, x, t) = \sum_{i=0}^{\infty} \frac{1}{\underline{|i}\,\underline{|i+1}} \mathsf{L}_i^{(1)}(x)\, x\, e^{-x}\, n_i(\mathbf{r}, t), \tag{4}$$

where $\mathsf{L}_n^{(1)}(x)$ is the generalised Laguerre polynomial of order unity and degree n and is given by

$$\mathsf{L}_n^{(1)}(x) = \frac{1}{x} e^{x} \frac{d^n}{d x^n} (e^{-x} x^{n+1}). \tag{5}$$

We have the following orthonormal property

$$\int_0^{\infty} \mathsf{L}_i^{(1)}(x)\, \mathsf{L}_k^{(1)}(x)\, x\, e^{-x}\, d x = \underline{|i}\,\underline{|i+1}\, \delta_{ik}. \tag{6}$$

Substituting (4) in (3) and multiplying the resulting equation by $\dfrac{1}{\underline{|k}\,\underline{|k+1}} \mathsf{L}_k^{(1)}(x)$ and integrating over x, we have

$$\sum_{i=0}^{\infty} \left(\frac{1}{\sqrt{x}}\right)_{ik} \frac{\partial n_i(\mathbf{r}, t)}{\partial t} - \frac{1}{3} l_{ik} \nabla^2 n_i(\mathbf{r}, t) + n_i(\mathbf{r}, t)\, (\Sigma_a)_{ik} + \gamma_{ik}\, n_i(r, t) = 0, \tag{7}$$

where

$$f_{ik}(x) = \frac{1}{\sqrt{\underline{|i}\,\underline{|i+1}}} \frac{1}{\sqrt{\underline{|k}\,\underline{|k+1}}} \int_0^{\infty} f(x)\, x\, e^{-x}\, \mathsf{L}_i^{(1)}(x)\, \mathsf{L}_k^{(1)}(x)\, d x, \tag{8}$$

and

$$\gamma_{ik} = \frac{1}{\sqrt{\underline{|i}\,\underline{|i+1}}} \frac{1}{\sqrt{\underline{|k}\,\underline{|k+1}}} \int_0^{\infty} \int_0^{\infty} d x\, d x' \, \{\Sigma_s(x \to x')\, x\, e^{-x}\, \mathsf{L}_i^{(1)}(x)\, \mathsf{L}_k^{(1)}(x) - \Sigma_s(x' \to x)\, x'\, e^{-x'}\, \mathsf{L}_i^{(1)}(x')\, \mathsf{L}_k^{(1)}(x)\}. \tag{9}$$

In Eq. (7) l denotes the transport mean free path and where we have omitted the suffix tr.

Let us now expand $n_i(\mathbf{r}, t)$ in terms of the eigenfunctions of the equation

$$\nabla^2 \chi_l(r) + B_l^2\, \chi_l(r) = 0,$$

where B_l^2 is the buckling corresponding to the lth harmonic. Now

$$n_i(\mathbf{r}, t) = \sum_l s_l^{(i)}(t)\, \chi_l(\mathbf{r}). \tag{10}$$

Since we are interested in times sufficiently long compared with the slowing-down time, all higher harmonics must have died out and we shall be left with only the fundamental mode. We then have

$$n_i(r, t) = s^{(i)}(t)\, \chi_0(\mathbf{r}), \tag{11}$$

where $\chi_0(\mathbf{r})$ is the eigenfunction corresponding to the lowest eigenvalue B^2.

3. Consideration of terms $i=0, 1$ and $k=0, 1$

In Eq. (7) we have an infinite set of simultaneous equations. As a first approximation we shall retain only terms corresponding to $i=0, 1$ and $k=0, 1$. We have then the following two simultaneous equations:

$$\left(\frac{1}{\sqrt{x}}\right)_{00}\frac{d\,s^{(0)}}{d\,t}+\left(\frac{1}{\sqrt{x}}\right)_{10}\frac{d\,s^{(1)}}{d\,t}+\frac{1}{3}\,l_{00}\,B^2\,s^{(0)}+s^{(0)}\,(\Sigma_a)_{00}+\frac{1}{3}\,l_{10}\,B^2\,s^{(1)}+s^{(1)}\,(\Sigma_a)_{10}=0 \qquad (12\text{ a})$$

for $k=0$ and $i=0$ and 1; and

$$\left(\frac{1}{\sqrt{x}}\right)_{01}\frac{d\,s^{(0)}}{d\,t}+\left(\frac{1}{\sqrt{x}}\right)_{11}\frac{d\,s^{(1)}}{d\,t}+\frac{1}{3}\,l_{01}\,B^2\,s^{(0)}+\frac{1}{3}\,l_{11}\,B^2\,s^{(1)}+s^{(0)}\,(\Sigma_a)_{01}+s^{(1)}\,(\Sigma_a)_{11}+\gamma_{11}\,s^{(1)}=0 \qquad (12\text{ b})$$

for $k=1$ and $i=0$ and 1.

Let the solutions of (12) be

$$\left.\begin{aligned} s^{(0)}&=e^{-\lambda t}\\ \text{and}\qquad s^{(1)}&=\alpha\,e^{-\lambda t}.\end{aligned}\right\} \qquad (13)$$

Substituting (13) in (12) we have

$$-\left(\frac{1}{\sqrt{x}}\right)_{00}\lambda-\left(\frac{1}{\sqrt{x}}\right)_{10}\lambda\,\alpha+\frac{1}{3}\,l_{00}\,B^2+(\Sigma_a)_{00}+\alpha\,(\Sigma_\alpha)_{10}+\frac{1}{3}\,l_{01}\,B^2\,\alpha=0 \qquad (14\text{ a})$$

and

$$-\left(\frac{1}{\sqrt{x}}\right)_{00}\lambda-\left(\frac{1}{\sqrt{x}}\right)_{11}\lambda\,\alpha+\frac{1}{3}\,l_{01}\,B^2+\frac{1}{3}\,l_{11}\,B^2\,\alpha+(\Sigma_\alpha)_{10}+\alpha\,(\Sigma_\alpha)_{11}+\alpha\,\gamma_{11}=0. \qquad (14\text{ b})$$

Eliminating α in the above two equations, we get the following quadratic equation in λ

$$\begin{aligned}\lambda^2+2\,\lambda\left[-K-\frac{4}{3\sqrt{\pi}}\gamma_{11}+\frac{4\,B^2}{9\sqrt{\pi}}\left(\frac{l_{01}}{\sqrt{2}}-\frac{7\,l_{00}}{8}-l_{11}\right)\right]+K^2+\frac{8}{3\sqrt{\pi}}\gamma_{11}\,K\\ -\frac{K B^2}{72\sqrt{\pi}}\left(\frac{l_{01}}{\sqrt{2}}-\frac{7}{8}\,l_{00}-l_{11}\right)+\frac{16}{9\,\pi}\,B^2\,l_{00}\,\gamma_{11}+\frac{16\,B^2}{27\,\pi}\,(l_{11}\,l_{00}-l_{01}^2)=0, \qquad (15)\end{aligned}$$

where we have made use of the following:

$$\left(\frac{1}{\sqrt{x}}\right)_{00}=\frac{\sqrt{\pi}}{2};\;\left(\frac{1}{\sqrt{x}}\right)_{10}=\frac{\sqrt{\pi}}{4\sqrt{2}}\;\text{ and }\;\left(\frac{1}{\sqrt{x}}\right)_{11}=\frac{7\sqrt{\pi}}{16}, \qquad (16)$$

which can be easily deduced using Eqs. (8) and (5). We also note that $f_{ik}=f_{ki}$. The macroscopic absorption cross section Σ_a is assumed to be inversely proportional to velocity *i.e.*, $\Sigma_a=\dfrac{K}{\sqrt{x}}$.

There exist two real roots of λ, of which we are interested in the smaller one. It is easy to show that if one retains terms up to order B^4, the smaller of the two roots is

$$\lambda = K + \frac{2\, l_{00}\, B^2}{3\sqrt{\pi}} - \frac{2\, B^4}{9\sqrt{\pi}\,\gamma_{11}} \left\{ l_{01} - \frac{l_{00}}{2\sqrt{2}} \right\}^2. \tag{17}$$

In proper units, λ is given by

$$\lambda = \Sigma_a v + \frac{2}{3}\frac{l_{00}\, v_0}{\sqrt{\pi}}\, B^2 - \frac{2\, B^4\, v_0}{9\sqrt{\pi}\,\gamma_{11}} \left(l_{01} - \frac{l_{00}}{2\sqrt{2}} \right)^2. \tag{18}$$

Now
$$\bar{D}_0 = \frac{2}{3}\frac{l_{00}\, v_0}{\sqrt{\pi}} = \frac{1}{3}\bar{l}_{tr}\,\bar{v}, \tag{19}$$

where $\bar{v}$ is mean velocity and is equal $\frac{2}{\sqrt{\pi}} v_0$ and $\bar{l}_{tr}$ is the average transport mean free path defined by

$$\bar{l}_{tr} = l_{00} = \int_0^\infty l_{tr}(x)\, \mathsf{L}_0^{(1)}(x)\, \mathsf{L}_0^{(1)}(x)\, e^{-x}\, x\, d\,x$$

$$= \int_0^\infty l_{tr}(x)\, x\, e^{-x}\, d\,x = \frac{1}{(k_0 T)^2} \int_0^\infty \lambda_{tr}(E)\, E \exp\left(-E/k_0 T\right) d\,E,$$

which is the same as A_0 of Eq. (9) of paper (I).

Also
$$l_{01} = \frac{1}{\sqrt{2}} \int_0^\infty l_{tr}(x)\, x\, e^{-x}\, \mathsf{L}_0^{(1)}(x)\, \mathsf{L}_1^{(1)}(x)\, d\,x$$

$$= \frac{1}{\sqrt{2}} \int_0^\infty l_{tr}(x)\, x\, e^{-x}\,(2 - x)\, d\,x$$

$$= \sqrt{2}\, A_0 - \frac{1}{\sqrt{2}} A_1, \tag{20}$$

where
$$A_i = \frac{1}{(k_0 T)^{i+2}} \int_0^\infty l_{tr}(E)\, E^{i+1} \exp\left(-E/k_0 T\right) d\,E. \tag{21}$$

It is shown in the appendix (Eq. A-20) that

$$4\,\gamma_{11} = M_2, \tag{22}$$

where M_2 is given by Eq. (4) of paper I. Using the above results (18) becomes

$$\lambda = \Sigma_a v + B^2\, \bar{D}_0 - B^4\, \bar{D}_0\, C \tag{23}$$

where
$$C=\frac{\sqrt{\pi}\,\bar{D}_0}{v_0\,M_2}\left(\frac{A_1}{A_0}-\frac{3}{2}\right)^2, \tag{24}$$

which is the same as equation (12) of paper I except that one of the $\bar{D}_0$'s here is taken with B^4. Thus if we retain terms corresponding to $i=0,1$ and $k=0,1$ in Eq. (7) we get the same result as was obtained earlier using the variational principle. To get a non-Maxwellian correction to the diffusion cooling constant C, we must carry out the next approximation, *i.e.*, retain terms corresponding to $i=0.\ 1,2$ and $k=0,1$ and 2. This will be done in Section 6.

4. Terms of order B^6 and B^8

Since the moderating assemblies have to be necessarily small for an accurate experimental determination of C, it is important to investigate the effect of terms of order B^6 and B^8 on the decay constant. Hitherto, experimenters have fitted their λ versus B^2 curve with a formula of the type (23), since neither the theoretical estimate of the higher order terms was known nor the experimental accuracy was sufficient. In deriving expressions for terms of order B^6 and B^8, we shall put $K=0$ in (15) *i.e.*, we consider a case with no absorption, since it simplifies the algebra and the results of interest are unaffected. The expression for the smaller of the two roots of λ can be put in the form

$$\lambda=\left[\frac{4}{3\sqrt{3}}\gamma_{11}-\frac{4B^2}{9\sqrt{\pi}}F\right]-\left[\frac{4}{3\sqrt{\pi}}\gamma_{11}-\frac{4B^2}{9\sqrt{\pi}}F\right]$$
$$\times\left[1-\left\{\frac{l_{00}B^2}{\gamma_{11}}+\frac{B^4}{3\gamma_{11}^2}(l_{00}l_{11}-l_{01}^2)\right\}\left\{1-\frac{B^2F}{3\gamma_{11}}\right\}^{-2}\right]^{\frac{1}{2}}, \tag{25}$$

where
$$F=\left(\frac{l_{01}}{\sqrt{2}}-\frac{7\,l_{00}}{8}-l_{11}\right). \tag{26}$$

It is now straightforward to show that terms of order B^6 and B^8 in λ are, respectively

$$-\frac{2B^6}{9\sqrt{\pi}\,\gamma_{11}^2}\left[\left(l_{11}+\frac{7}{8}l_{00}-\frac{l_{01}}{\sqrt{2}}\right)\left(\frac{11}{24}l_{00}^2+\frac{1}{3\sqrt{2}}l_{00}l_{01}-\frac{1}{3}l_{01}^2\right)\right.$$
$$\left.-\frac{3}{8}l_{00}^3+\frac{1}{2}l_{00}l_{01}^2-\frac{1}{2}l_{00}^2l_{11}\right], \tag{27}$$

and

$$-\frac{2B^8}{27\sqrt{\pi}\,\gamma_{11}^3}\left[-\frac{1}{3}l_{00}F^3-F^2\left(\frac{3}{2}l_{00}^2+\frac{1}{3}(l_{00}l_{11}-l_{01}^2)\right)+\right.$$
$$\left.+l_{00}F\left(-\frac{15}{8}l_{00}^2-\frac{3}{2}(l_{00}l_{11}-l_{01}^2)\right)-\frac{45}{64}l_{00}^4-\frac{9}{8}l_{00}^2(l_{00}l_{11}-l_{01}^2)-\frac{1}{2}(l_{00}l_{11}-l_{01}^2)^2\right]. \tag{28}$$

In order to estimate the magnitude of these correction terms we shall assume that l_{tr} is constant. In that case, it is easy to show that

$$l_{01}=0,\quad l_{00}=l \quad \text{and} \quad l_{11}=l \quad \text{and} \quad F=-\tfrac{15}{8}\,l, \tag{29}$$

where l stands for the transport mean free path and corresponds to A_0 of paper I. Equations (27) and (28) then, respectively, reduce to

$$-\frac{2\,B^6}{9\sqrt{\pi}\,\gamma_{11}^2}\left[-\frac{1}{64}\right] l^3, \tag{30}$$

and

$$-\frac{4\,B^8}{3\sqrt{\pi}\,\gamma_{11}^3}\,\frac{1}{128}\left(-\frac{1}{72}\right) l^4. \tag{31}$$

In the same approximation, the term of order B^4 from (17) is

$$-\frac{2\,B^4}{9\sqrt{\pi}\,\gamma_{11}}\,\frac{l^2}{8}. \tag{32}$$

Adding (30), (31) and (32) and using (19) we have for the diffusion cooling constant the expression

$$C=\left[1-\frac{B^2\,l}{8\,\gamma_{11}}-\frac{2\,B^4\,l^2}{128\times 3\,\gamma_{11}^2}\right]\frac{\sqrt{\pi}\,\bar{D}_0}{16\,\gamma_{11}\,v_0}. \tag{33 a}$$

The smallest assembly used by deSaussure and Silver in their experiments on beryllium had $B^2=7\times10^{-2}\ \text{cm}^{-2}$. Remembering that $l=1.51$ cm for beryllium and $4\,\gamma_{11}=M_2=0.32\ \text{cm}^{-1}$ (values from paper I), we find that the second and third terms in the square brackets in (33 a) contribute nearly 16 per cent and 1 per cent, respectively. It is enough, therefore, to take only terms up to order B^6. It should be pointed out that the correction again depends sensitively on the values of l_{ik}. The reason we have used the constant transport mean free path approximation in estimating the magnitude of these corrections is that we get a better agreement with the experimental value of C of deSaussure and Silver.

It is instructive to note that the above corrections at low temperatures are temperature sensitive, whereas at high temperatures they remain constant for a given value of B^2. At low temperatures l increases and γ_{11} decreases and, therefore, the correction term increases rapidly in magnitude. To reduce this correction the assembly has, therefore, to be very large at low temperatures.

Since the main correction to C is due to the term of order B^6, we shall now investigate it in the next higher approximation *i.e.*, we now retain the first three terms in the Laguerre expansion and shall again assume that the transport mean free path is constant. We shall give only the final result. Instead of (33 a) we now get

$$C=\frac{\sqrt{\pi}\,\bar{D}_0}{16\,\gamma_{11}\,v_0}\left[1-\frac{1}{8}\,\frac{B^2\,l}{\gamma_{11}}\left(1-\frac{3}{4}\,\frac{\gamma_{11}}{\gamma_{22}}+\frac{57}{192}\,\frac{\gamma_{11}^2}{\gamma_{22}^2}\right)\right], \tag{33 b}$$

where we have neglected terms containing γ_{12} since they are very small. On comparing (33 b) with (33 a), it is evident that the correction due to the B^6 term is reduced by the factor $\left(1-\frac{3}{4}\frac{\gamma_{11}}{\gamma_{22}}+\frac{57}{192}\left(\frac{\gamma_{11}}{\gamma_{22}}\right)^2\right)$, which is nearly 7/10 if we take the value $\gamma_{22}=2\gamma_{11}$ (see Appendix). Thus, the 16 per cent correction as estimated before will now be reduced to only 11 per cent. Therefore, even for the smallest assembly of deSaussure and Silver, the correction due to terms of order B^6 is only 50 per cent of the non-Maxwellian correction. And on the average it would not amount to more than five to six per cent. Hence the main correction to the diffusion cooling constant is the one which is non-Maxwellian.

5. Estimation of the mean energy of neutrons inside and outside the assembly

The mean energy per neutron is given by

$$\bar{x}=\frac{\int\limits_0^\infty x\,\phi(\mathbf{r},x,t)\,dx}{\int\limits_0^\infty \phi(\mathbf{r},x,t)}=2-\sqrt{2}\,\frac{n_1(\mathbf{r},t)}{n_0(\mathbf{r},t)},$$

$$=2-\sqrt{2}\cdot\alpha, \tag{34}$$

where in deriving (34) we have used (4) and (13).

For the value of λ under consideration, α from equation (14), if we retain terms up to order B^4, is given by

$$\alpha=-\frac{B^2}{3\gamma_{11}}\left(l_{01}-\frac{l_{00}}{2\sqrt{2}}\right)+\frac{B^4}{18\sqrt{2}}\frac{1}{\gamma_{11}^2}\left(l_{01}-\frac{l_{00}}{2\sqrt{2}}\right)^2, \tag{35 a}$$

which for constant l_{tr} reduces to

$$\alpha=+\frac{B^2 l}{6\sqrt{2}\gamma_{11}}+\frac{B^4}{18\sqrt{2}}\frac{1}{\gamma_{11}^2}\cdot\frac{l^2}{8}. \tag{35 b}$$

Using the above value of α in (34) we have

$$\bar{x}=2\left[1-\frac{B^2 l}{12\gamma_{11}}-\frac{B^4 l^2}{18}\frac{1}{16\gamma_{11}^2}\right]. \tag{36}$$

Thus for $B^2=0$, $\bar{x}=2$, as one would expect. If we now substitute the values for B^2, l and γ_{11} of the previous section, we find that the last two terms in the square brackets in (36) contribute nearly 13 per cent. Therefore, in a finite beryllium assembly with $B^2=7\times10^{-2}$ cm^2, the inside mean energy per neutron is 13 per cent less than what it would be in an infinite assembly. Again this deviation would increase rapidly at low temperatures.

We shall now estimate the mean energy per neutron in the leaking-out flux, since it is this quantity which is usually measured. The flux outside is given by

$$\phi_{\text{out}} = \frac{l_{tr}(x)}{3} \nabla \phi(r, x, t),$$

or

$$\bar{x}_{\text{out}} = \frac{1}{3} \int_0^\infty l_{tr}(x) \cdot x \nabla \phi \, dx \Bigg/ \frac{1}{3} \int_0^\infty l_{tr}(x) \nabla \phi \, dx.$$

Using (4) and (13) we have

$$\bar{x}_{\text{out}} = \frac{\int_0^\infty l_{tr}(x) x^2 e^{-x} \mathsf{L}_0^{(1)}(x) \, dx + \frac{\alpha}{\sqrt{2}} \int_0^\infty x \cdot l_{tr}(x) x e^{-x} \mathsf{L}_1^{(1)}(x) \, dx +}{\int_0^\infty l_{tr}(x) x e^{-x} \mathsf{L}_0^{(1)}(x) \, dx + \frac{\alpha}{\sqrt{2}} \int_0^\infty l_{tr}(x) x e^{-x} \mathsf{L}_1^{(1)}(x) \, dx + \cdots}$$

$$\simeq \frac{\int_0^\infty l_{tr}(x) x^2 e^{-x} \, dx + \frac{\alpha}{\sqrt{2}} \int_0^\infty l_{tr}(x) x^2 e^{-x} (2 - x) \, dx}{\int_0^\infty l_{tr}(x) x e^{-x} \, dx + \frac{\alpha}{\sqrt{2}} \int_0^\infty l_{tr}(x) x e^{-x} (2 - x) \, dx}$$

$$= A_1 + \frac{\alpha}{\sqrt{2}} \cdot (2A_1 - A_2) \Bigg/ A_0 + \frac{\alpha}{\sqrt{2}} (2A_0 - A_1), \tag{37}$$

where A_i it given by Eq. (21). Substituting the value of α from (35a) and retaining terms up to B^2, we have

$$\bar{x}_{\text{out}} = \frac{A_1}{A_0} \left[1 - \frac{2}{3} \frac{B^2 A_0}{M_2} \left(\frac{A_1}{A_0} - \frac{3}{2} \right) \left(\frac{A_2}{A_1} - \frac{A_1}{A_0} \right) \right]. \tag{38}$$

Thus for

$$B^2 = 0, \quad (\bar{x})_{\text{out}} = \frac{A_1}{A_0}.$$

$$= 2,$$

if the transport mean free path is taken to be constant. In the case of a beryllium assembly with $B^2 = 7 \times 10^{-2}$ cm², the correction term in (38) is of the order of 10 per cent for constant l_{tr}. Therefore for the smallest assemblies considered the mean energy per neutron is nearly 10 per cent less than what it would be for an infinite assembly.

Now the mean energy inside the assembly to order B^2 is

$$\bar{x}_{\text{in}} \quad = 2\left[1 - \frac{2}{3}\frac{B^2 A_0}{M_2}\left(\frac{A_1}{A_0} - \frac{3}{2}\right)\right].$$

Therefore,

$$\bar{x}_{\text{in}} - \bar{x}_{\text{out}} = \left(2 - \frac{A_1}{A_0}\right) + \frac{2}{3}\frac{B^2}{M_2}\left(\frac{A_1}{A_0} - \frac{3}{2}\right)\left\{A_1\left(\frac{A_2}{A_1} - \frac{A_1}{A_0}\right) - 2A_0\right\}, \tag{39 a}$$

$$\simeq 2 - \frac{A_1}{A_0}. \tag{39 b}$$

If we assume that l_{tr} is independent of energy, $A_2 = 6A_0$ and $A_1 = 2A_0$, then from (39 b), $\bar{x}_{\text{in}} - \bar{x}_{\text{out}} = 0$, *i.e,* a measurement of the mean energy outside the assembly gives also the inside mean energy. But in reality l_{tr} depends on neutron energy and the difference of the energies inside and outside will be given approximately by Eq. (39 b). For beryllium this difference is nearly 7 per cent. However for graphite where $\frac{A_1}{A_0}$ is almost equal to 2, the difference is insignificant. If the moderating assembly is very small, the term involving B^2 in (39 a) should be taken into account.

6. Non-Maxwellian correction

In calculating the non-Maxwellian correction to the diffusion cooling constant, we have to retain terms at least up to $i = 2$ and $k = 2$ in the Laguerre expansion. We shall neglect absorption since it does not influence the result we are interested in and the algebra is considerably simplified. Putting $p_{ik} \equiv \left(\frac{1}{\sqrt{x}}\right)_{ik}$ and remembering that $p_{ik} = p_{ki}$, Eq. (7) becomes

$$\sum_{i=0}^{\infty} p_{ik}\frac{d s^{(i)}}{d t} + \frac{1}{3} l_{ik} B^2 S^{(i)} + \gamma_{ik} s^{(i)} = 0, \tag{40}$$

where we have made use of Eq. (11). For the case under consideration Eq. (40) gives the following three similtaneous equations:

$$p_{00}\frac{d s^{(0)}}{d t} + p_{10}\frac{d s^{(1)}}{d t} + p_{20}\frac{d s^{(2)}}{d t} l_{00} B^2 s^{(0)}$$

$$+ \frac{1}{3} l_{10} B^2 s^{(1)} + \frac{1}{3} l_{20} B^2 s^{(2)} = 0, \tag{41a}$$

$$p_{01}\frac{d s^{(0)}}{d t} + p_{11}\frac{d s^{(1)}}{d t} + p_{21}\frac{d s^{(2)}}{d t} + \frac{1}{3} l_{01} B^2 s^{(0)}$$

$$+ \frac{1}{3} l_{11} B^2 s^{(1)} - \frac{1}{3} l_{21} B^2 s^{(2)} + \gamma_{11} s^{(1)} + \gamma_{21} s^{(2)} = 0, \tag{41 b}$$

and
$$p_{02}\frac{d\,s^{(0)}}{d\,t}+p_{12}\frac{d\,s^{(1)}}{d\,t}+p_{22}\frac{d\,s^{(2)}}{d\,t}+\frac{1}{3}\,l_{02}\,B^2\,s^{(0)}$$
$$+\frac{1}{3}\,l_{12}\,B^2\,s^{(1)}+\frac{1}{3}\,l_{22}\,B^2\,s^{(2)}+\gamma_{12}\,s^{(1)}+\gamma_{22}\,s^{(2)}=0. \qquad (41\text{ c})$$

Let the solutions be

$$\left.\begin{aligned} s^{(0)}&=\delta\,e^{-\lambda t},\\ s^{(1)}&=\alpha\,e^{-\lambda t},\\ s^{(2)}&=\beta\,e^{-\lambda t}.\end{aligned}\right\} \qquad (42)$$

and

Substituting (42) in (41) we have

$$\left(-p_{00}\,\lambda+\frac{1}{3}\,l_{00}\,B^2\right)\delta-\alpha\left(p_{10}\,\lambda-\frac{1}{3}\,l_{01}\,B^2\right)-\beta\left(p_{20}\,\lambda-\frac{1}{3}\,l_{20}\,B^2\right)=0, \qquad (43\text{ a})$$

$$\left(-p_{01}\,\lambda+\frac{1}{3}\,l_{01}\,B^2\right)\delta-\alpha\left(p_{11}\,\lambda-\frac{1}{3}\,l_{11}\,B^2-\lambda_{11}\right)-\beta\left(p_{21}\,\lambda-\frac{1}{3}\,l_{21}\,B^2-\gamma_{21}\right)=0, \qquad (43\text{ b})$$

and

$$\left(-p_{02}\,\lambda+\frac{1}{3}\,l_{02}\,B^2\right)\delta-\alpha\left(p_{12}\,\lambda-\frac{1}{3}\,l_{12}\,B^2-\gamma_{12}\right)-\beta\left(p_{22}\,\lambda-\frac{1}{3}\,l_{22}\,B^2-\gamma_{22}\right)=0. \qquad (43\text{ c})$$

In order that there may exist a non-trivial solution for δ, α and β, the determinant formed by their coefficients in (43) must vanish. We would then get a cubic equation in λ. Making use of the following:

$$\left.\begin{aligned} &p_{00}=\frac{\sqrt{\pi}}{2};\quad p_{10}=\frac{\sqrt{\pi}}{4\sqrt{2}};\quad p_{11}=\frac{7\sqrt{\pi}}{16}\\ &\text{(see Eq. 16), and}\\ &p_{02}=\frac{\sqrt{3\,\pi}}{16},\quad p_{12}=\sqrt{\frac{\pi}{24}}\cdot\frac{15}{16}\quad\text{and}\quad p_{22}=\frac{51\sqrt{\pi}}{128},\end{aligned}\right\} \qquad (44)$$

which relations are easy to prove and remembering that $p_{ik}=p_{ki}$ we get the following equation:

$$\lambda^3+a\,\lambda^2+b\,\lambda+c=0, \qquad (45)$$

where

$$a=\frac{\pi}{16}\cdot\frac{256}{15\,\pi^{3/2}}\left[\frac{1}{3}\,B^2\left\{\frac{\sqrt{3}}{4}\,l_{02}+\frac{2\sqrt{3}}{\sqrt{2}}\,l_{12}+\frac{9}{4\sqrt{2}}\,l_{01}-\frac{141}{64}\,l_{00}\right.\right.$$
$$\left.\left.-3\,l_{11}-3\,l_{22}\right\}-3\,\gamma_{11}+\frac{2\sqrt{3}}{\sqrt{2}}\,\gamma_{12}-3\,\gamma_{22}\right], \qquad (46)$$

$$
\begin{aligned}
b = \frac{\sqrt{\pi}}{8}\,\frac{256}{15\,\pi^{3/2}} \Bigg[& \frac{1}{9} B^4 \Bigg\{ \frac{4}{\sqrt{2}} l_{01} l_{12} - \sqrt{3}\, l_{02} l_{11} + \frac{15}{\sqrt{24}} l_{01} l_{02} \\
& - \frac{7}{2} l_{02}^2 + \sqrt{3}\, l_{01} l_{12} - 4\, l_{12}^2 - \frac{15}{\sqrt{24}} l_{00} l_{12} + 4\, l_{11} l_{22} \\
& \frac{7}{2} l_{00} l_{22} - \frac{4}{\sqrt{2}} l_{01} l_{22} - \frac{51}{16} l_{01}^2 + \frac{51}{16} l_{00} l_{11} \Bigg\} \\
& + \frac{1}{3} B^2 \Bigg\{ \gamma_{12} \left(\sqrt{3}\, l_{01} + \frac{4}{\sqrt{2}} l_{02} - 8\, l_{12} - \frac{15}{\sqrt{24}} l_{00} \right) \\
& + \gamma_{11} \left(- \sqrt{3}\, l_{02} + 4\, l_{22} + \frac{51}{16} l_{00} \right) + \gamma_{22} \Bigg(4\, l_{11} + \frac{7}{2} l_{00} \\
& - \frac{4}{\sqrt{2}} l_{01} \Bigg) \Bigg\} + 4 \left(\gamma_{11} \gamma_{22} - \gamma_{12}^2 \right) \Bigg],
\end{aligned}
\tag{47}
$$

and

$$
\begin{aligned}
c = - \frac{256}{15\,\pi^{3/2}} \Bigg[& \frac{1}{27} B^6 \{ l_{02} (l_{01} l_{12} - l_{02} l_{11}) - l_{12} (l_{00} l_{12} \\
& - l_{01} l_{02}) - l_{22} (l_{01}^2 - l_{00} l_{11}) \} + \frac{1}{9} B^4 \{ 2\, \gamma_{12} (l_{01} l_{02} \\
& - l_{00} l_{12}) + \gamma_{11} (l_{00} l_{22} - l_{02}^2) + \gamma_{22} (l_{00} l_{11} - l_{01}^2) \} \\
& + \frac{1}{3} B^2 \{ l_{00} \gamma_{11} \gamma_{22} - l_{00} \gamma_{12}^2 \} \Bigg].
\end{aligned}
\tag{48}
$$

To determine λ we proceed as follows:

We know that the approximate value of λ which satisfies (45) is λ_1, and it is given by Eq. (18) without the first term since in this case we have taken $\Sigma_a = 0$. We write λ_1 as

$$\lambda_1 = \Delta (1 - \varepsilon), \tag{49}$$

where

$$\Delta = \frac{2\, l_{00} B^2}{3 \sqrt{\pi}}, \tag{50}$$

and

$$\varepsilon = \frac{B^2}{3\, \gamma_{11} l_{00}} \left(l_{01} - \frac{l_{00}}{2 \sqrt{2}} \right)^2. \tag{51}$$

Let the value of λ, which satisfies Eq. (45) be $\lambda_1 + x$, where x is small. Then $\lambda_1 + x$ is one of the roots of Eq. (45) in which we are interested. x is then given by

$$x = - \frac{\lambda_1^3 + a\, \lambda_1^2 + b\, \lambda_1 + c}{3\, \lambda_1^2 + 2\, a\, \lambda_1 + b}, \tag{52}$$

where we have retained only quantities of the order (x/λ_1). It is now a straightforward matter, though somewhat lengthy, to determine x. We expand all terms in the numerator and denominator of (52) in powers of B^2. We then have

$$x = -\frac{2B^4}{9\sqrt{\pi}\,\gamma_{11}}\left(l_{01}-\frac{l_{00}}{2\sqrt{2}}\right)^2 - \frac{2\,l_{00}B^2}{3\sqrt{\pi}}\left[\frac{1}{3}\frac{B^2F_1}{l_{00}(\gamma_{12}^2-\gamma_{11}\gamma_{22})}\right.$$
$$\left. + \frac{B^2F_2}{12(\gamma_{12}\gamma_{22}-\gamma_{12}^2)} + \frac{l_{00}B^2}{12(\gamma_{11}\gamma_{22}-\gamma_{12}^2)}(-3\gamma_{11}+\sqrt{6}\,\gamma_{12}-3\gamma_{22})\right], \quad (53)$$

where $$F_1 = 2\gamma_{12}(l_{01}l_{02}-l_{00}l_{12}) + \gamma_{11}(l_{00}l_{22}-l_{02}^2) + \gamma_{22}(l_{00}l_{11}-l_{01}^2), \quad (54)$$

and

$$F_2 = \gamma_{12}\left(\sqrt{3}\,l_{01}+\frac{4}{\sqrt{2}}l_{02}-8\,l_{12}-\frac{15}{\sqrt{24}}l_{00}\right)+$$
$$\gamma_{11}\left(-\sqrt{3}\,l_{02}+4\,l_{22}+\frac{51}{16}l_{00}\right)+\gamma_{22}\left(4\,l_{11}+\frac{7}{2}l_{00}-\frac{4}{\sqrt{2}}l_{01}\right). \quad (55)$$

In deriving (53) terms only up to order B^4 have been retained. It is shown in the appendix that for a heavy monoatomic gas $\gamma_{22}=2\gamma_{11}$ and $\gamma_{12}=0$. For an actual solid this will, however, be approximately true. Assuming that $\gamma_{12}\ll\gamma_{11}$ and $\gamma_{12}\ll\gamma_{22}$, we get a very simple expression for x and therefore for λ. Hence

$$\lambda = \frac{2\,l_{00}B^2}{3\sqrt{\pi}} - \frac{2B^4}{9\sqrt{\pi}\,\gamma_{11}}\left(l_{01}-\frac{1}{2\sqrt{2}}l_{00}\right)^2 - \frac{2B^4}{9\sqrt{\pi}\,\gamma_{22}}\times\left(l_{02}-\frac{\sqrt{3}}{8}l_{00}\right)^2$$
$$= \frac{2\,l_{00}B^2}{3\sqrt{\pi}} - \frac{2B^4}{9\sqrt{\pi}\,\gamma_{11}}\left(l_{01}-\frac{1}{2\sqrt{2}}\right)^2\left[1+\frac{\gamma_{11}}{\gamma_{22}}\frac{\left(l_{02}-\frac{\sqrt{3}}{8}l_{00}\right)^2}{\left(l_{01}-\frac{1}{2\sqrt{2}}l_{00}\right)^2}\right]. \quad (56)$$

The second term in the square brackets in (56) represents the main term of the non-Maxwellian correction to the diffusion cooling constant, arising as a result of considering the first three terms in the Laguerre expansion for the neutron-velocity distribution function. It represents an improvement over the result deduced from the variational principle in paper I. We shall now estimate this correction which we call $\Delta C/C$ for the case of constant transport mean free path.

$$\frac{\Delta C}{C} = \frac{\gamma_{11}}{\gamma_{22}}\cdot\frac{(\sqrt{3/8})^2\cdot \bar{l}_{tr}^2}{\left(\frac{1}{2\sqrt{2}}\right)^2\bar{l}_{tr}^2}$$
$$\simeq \frac{3}{16}$$

i.e., $\Delta C/C$ is nearly 19 per cent.

One might conclude that the use of a Maxwellian trial function in the variational approach of paper I underestimates the diffusion cooling constant by nearly 20 per cent. Since the error in C is of second order in the error in the spectrum, the deviations from the equilibrium neutron spectrum cannot be described in terms of a neutron temperature which differs from the moderate temperature.

It is not difficult, though somewhat tedious, to estimate the correction due to terms higher than the third in the Laguerre expansion. We have not done so but our guess is that these terms may contribute nearly five to six per cent. Thus the total non-Maxwellian correction to C is nearly 25 per cent. The higher order corrections can be easily estimated on the basis of a heavy gas model.

7. Variation of the diffusion cooling constant with temperature

The variation of the diffusion cooling constant with the temperature of the moderating assembly is of special physical interest in the range of low temperatures *i.e.*, $T \ll \theta$, θ being the Debye temperature of the solid, since it is in this range that the effect of crystalline binding plays a dominant role. The region of high temperatures *i.e.*, $T \gtrsim \theta$ is not of much physical interest, though it may be of some practical importance. As far as the author is aware, no reliable experimental results concerning the variation of the diffusion cooling constant with temperature are available. deSaussure and Silver (1958) are planning to do these experiments in a beryllium assembly in a fairly wide temperature range. Their very preliminary experiment at 100°K indicates that the neutron decay does not become exponential even after a fairly long interval of time This obviously means that the time constant of the thermalization process is very large at low temperatures, as expected from the theory.

It is not possible to derive an analytical expression showing an explicit temperature dependence of the diffusion cooling constant. We shall, therefore, consider here the two limiting cases: (i) $\theta/T \to \infty$ and (ii) $\theta/T \to 0$, the formulae for which have been derived in the appendix.

Case (i) $\theta/T \to \infty$.

From Eq. (A. 19)

$$\bar{D}_0 \propto \left(\frac{\theta}{T}\right)^3 \cdot T^{\frac{1}{2}},$$

and from Eq. (A. 13)

$$M_2 \propto \left(\frac{T}{\theta}\right)^3.$$

And since $C = \frac{\sqrt{\pi}}{4} \frac{\bar{D}_0}{v_0 M_2}$, we have

$$C \propto \left(\frac{\theta}{T}\right)^3 T^{\frac{1}{2}} \Big/ T^{\frac{1}{2}} \left(\frac{T}{\theta}\right)^3,$$

or

$$C \propto \left(\frac{\theta}{T}\right)^6. \tag{57}$$

Thus the diffusion cooling constant would increase very rapidly with the decrease of temperature for low temperatures of the moderating assembly. This

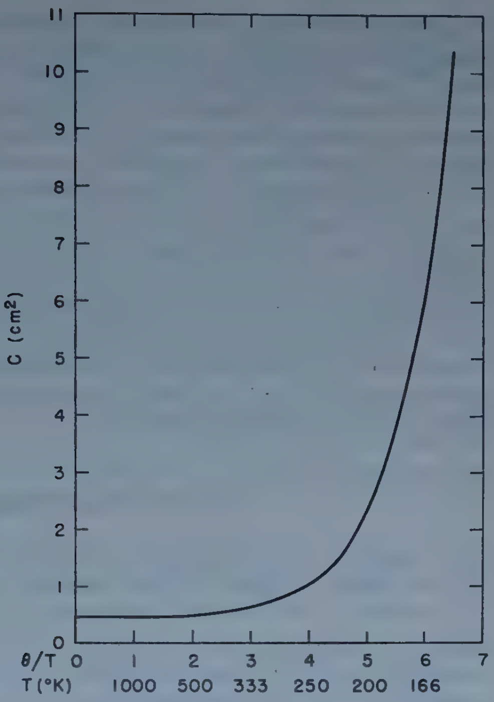

Fig. 1. Diffusion cooling constant C $\left(=0.443\,\frac{\bar{D}_0}{v_0 M_2}\right)$ as a function of θ/T for beryllium.

would imply that the leakage rate would be so large that probably a thermal equilibrium would never be attained at these temperatures.

Case (ii) $\theta/T \to 0$.

$$\bar{D}_0 = \frac{1}{3}\,\bar{l}_{tr}\,\bar{v}$$

$$= \text{const } T^{\frac{1}{2}},$$

and from Eq. (A. 12) $$M_2 = \text{const.}$$

Therefore, $$C = \text{constant.} \tag{58}$$

Equation (58) will probably be true even for $T \simeq \theta$. Detailed calculations in the case of beryllium indicate that (58) is true in the range $\theta/T = 0$ to $\theta/T = 2$. It is necessary to make some comments here regarding equation (57). Equation (57) is true only in the limit $\left(\frac{\theta}{T}\right) \to \infty$. In deriving (57), it has been assumed that almost all the contribution to $\bar{D}_0$ arises from the inelastic scattering of neutrons by the lattice, which would not be true in general for all solids except at very low temperatures. However, in the case of beryllium even at 150°K, the foregoing assumption may be true, since the Brag cutoff for beryllium is as high as 60°K and as we know, below the Bragg cutoff there is no elastic scattering. We might, therefore, expect Eq. (57) to hold for beryllium at 150°K and below. On the other hand, in the case of graphite, whose Bragg cutoff is nearly 20°K, our assumption is probably not true and Eq. (57) will not hold at these temperatures. Here most of the contribution to $\bar{D}_0$ would still probably come from elastic scattering, which is not temperature sensitive. In that case $\bar{D}_0 \propto T^{\frac{1}{2}}$ and $M_2 \propto (T/\theta)^3$ as before and, therefore, $C \propto (\theta/T)^3$.

In Fig. 1 the diffusion cooling constant for beryllium is plotted as a function of θ/T. These calculations are exact. The non-Maxwellian correction as given by (56), and which is nearly $+20$ per cent, has not been added. It may be noted that as (θ/T) increases, M_2 decreases and therefore the correction due to terms of order B^6 as given by (33) also increases and it has an opposite sign to that of the non-Maxwellian correction. From the figure it is clear that from $(\theta/T)=0$ to $(\theta/T)=2$, C almost remains constant; it then slowly increases and after $(\theta/T)=4$ it starts increasing rapidly. It would be interesting to verify these results experimentally.

8. Non-diffusion correction

In Section 3 we have, in the diffusion approximation, derived an expression for the decay constant of the fundamental mode in a multi-velocity theory. It is, however, well known that even in a one-velocity group theory the decay constant would contain terms of order B^4 and higher, which ordinarily are negligible for very large assemblies. As we are mainly concerned here with small assemblies for the determination of C, it would be necessary to estimate the magnitude of these terms. We might call this correction the non-diffusion correction, for the simple reason that no diffusion approximation is made and the transport equation is solved under certain approximations. We shall neither give here any derivation of the results nor the assumptions under which it is derived, since it is essentially contained in Davisson's book (1957) (see also Sjöstrand, 1959).

We start with Eq. (5.5) of Davisson:

$$\frac{1}{C} = \frac{L}{2l} \log \frac{L+l}{L-l}, \tag{59}$$

where

$$l^{-1} = \int_0^\infty \left(\Sigma_s(v) - \frac{\gamma}{v}\right) F(v)\, dv \cdot v \Big/ \int_0^\infty v\, F(v)\, dv = \bar{\Sigma}_s - \frac{\gamma}{\bar{v}}; \tag{60}$$

$$C = \bar{\Sigma}_s \Big/ \bar{\Sigma}_s - \frac{\gamma}{\bar{v}}, \tag{61}$$

and

$$\frac{1}{iL} = B, \tag{62}$$

where B^2 is the geometrical buckling.

The decay constant λ is given by

$$\lambda = \gamma + v\, \Sigma_a. \tag{63}$$

$\bar{\sum}_s$ in (61) is the macroscopic scattering cross section averaged over the neutron spectrum $F(v)$ of Eq. (4.12) of Davisson. In the case of thermal equilibrium it is given by the Maxwell distribution.

From Eq. (59)–(63), it is easy to show that

$$\lambda = v \sum_a + \bar{v} \bar{\sum}_s \left(1 - \frac{B/\bar{\sum}_s}{\tan B/\bar{\sum}_s}\right) = v \sum_a + \frac{\bar{v} B^2}{3 \bar{\sum}_s} \left(1 + \frac{B^2}{15 \bar{\sum}_s^2} + \cdots\right). \tag{64}$$

In deducing (64), the scattering is assumed to be isotropic in the laboratory system. The anisotropy in scattering can be taken care of as in Section 17.3 of Davisson's book. For the moderators of interest like beryllium, beryllium oxide and graphite, it is sufficient to consider terms of order $\left(\frac{m}{M}\right)$ in the differential scattering cross section. In that case one need retain only the P_1 term in the Legendre expansion of the differential scattering cross section. We can then replace $\bar{\sum}_s$ in (64) by $\sum_{tr} = \bar{\sum}_s (1 - \overline{\cos\theta})$, where $\sum_{tr}$ is the average macroscopic transport cross section. Equation (64) then becomes

$$\lambda = v \sum_a + \bar{v} \frac{B^2}{3 \sum_{tr}} \left(1 + \frac{B^2}{15 \sum_{tr}^2} + \cdots\right) = v \sum_a + B^2 \bar{D}_0 \left(1 + \frac{B^2}{15} \bar{l}_{tr}^2 + \cdots\right). \tag{65}$$

Since $\bar{l}_{tr}$ is of the order of unity, it is seen that even for assemblies for which $B^2 = 7 \times 10^{-2}$ cm^2, the non-diffusion correction term is nearly one per cent or so.

From Eq. (18) we see that the multi-velocity correction in the constan transport mean free path approximation is $\frac{B^2}{6 M_2} \bar{l}_{tr}$. The ratio of the multi-velocity to the non-diffusion correction is therefore,

$$\frac{(\Delta \lambda) \text{ multi-velocity}}{(\Delta \lambda) \text{ non-diffusion}} = \frac{15}{6} \cdot \frac{1}{\bar{l}_{tr} M_2}, \tag{66}$$

which in the case of beryllium is nearly 5. Thus one can always neglect the non-diffusion correction in comparison to the multi-velocity correction to the decay constant.

9. Results and discussion

Before we proceed to compare the theoretical values of the diffusion cooling constant with the experimental values, it is necessary to make a few general remarks. For an accurate experimental determination of the diffusion cooling constant, the moderator assemblies have to be necessarily small, and in that case, according to formula (33), terms of order B^6 in the expression for the decay constant should be taken into account. This correction is negligible only if $B^2 \bar{l}_{tr} \ll 8 \gamma_{11}$ or $2 M_2$. Hitherto, experimental curves for the decay constant have been fitted with the relation of the type $\lambda = \sum_a v + \bar{D}_0 B^2 - C \bar{D}_0 B^4$, thus neglecting terms of order B^6.

In calculating the numerical values of the diffusion cooling constant from our formula, we shall assume that the transport mean free path remains constant with energy, since this assumption gives a much better agreement with experiment. In fact the transport mean free path, in the region of Bragg peaks varies very violently with neutron energy, but in the process of averaging over a Maxwell distribution corresponding to the temperature of the moderator, these fluctuations are smoothed out. Detailed calculations of Singwi and Kothari (1958) show that the above assumption is fairly good in the case of graphite but not so good in the case of beryllium and beryllium oxide. With our present knowledge of the vibrational spectra of these solids and with the use of the incoherent approximation, it is not possible to calculate the quantities l_{00}, l_{01}, l_{11}, etc. with an accuracy of more than five to ten per cent which, however, is not enough for the calculation of quantities like

$$\left(l_{01}-\frac{l_{00}}{2\sqrt{2}}\right)^2 \quad\text{and}\quad \left(l_{02}-\frac{\sqrt{3}}{8}l_{00}\right)^2$$

in formula (56). We shall now consider the cases of beryllium, beryllium oxide and graphite separately.

(a) *Beryllium.*—Here we shall consider only the more recent measurements of deSaussure and Silver (1958). Their value of the diffusion cooling constant C is 1.1 ± 0.9 cm^2 and the maximum value of B^2 in their experiment was 7×10^{-2} cm^2. For this value of B^2, the correction term of order B^6 in (33 b) is -11 %, and the non-Maxwellian correction of formula (56) is nearly $+20$ %. If, however, we assume that on the average the former correction is of the order of five per cent, the net correction to the diffusion cooling constant is $+15$ %. Thus the calculated value $(C)_{\mathrm{cal}}$ is 0.9 cm^2 (with $\bar{D}_0=1.25\times10^5$ cm^2 sec^{-1}), whereas the experimental value, $(C)_{\mathrm{exp}}$ is 1.1 ± 0.9 cm^2. The agreement between the two values is fairly good. It would further improve by five per cent or so if we add higher order non-Maxwellian corrections. Whether this agreement between theory and experiment is significant is questionable for two reasons: in the first place the experimental error is very large and secondly we have assumed that the transport mean free path is independent of energy. The experimental value of C of Antonov *et al.* (1956) is 2.5 ± 0.8 cm^2. In the calculations the value of M_2 as given in paper I has been used. Our value of M_2 differs slightly from Nelkin's value which is 0.36 cm^{-1}, since he has used for the Debye temperature for beryllium a value of 930°K whereas we have taken it to be 1000°K.

(b) *Beryllium oxide.*—So far as this moderator is concerned, the only available experimental value of C is that of Iyengar *et al.* (1957). Their value of $C\bar{D}_0$ is 3.85×10^5 cm^4 sec^{-1}. The maximum value of B^2 in their experiment was 6.0×10^{-3} cm^2. Using the theoretical values of $\bar{l}_{tr}=1.58$ cm (Singwi and Kothari (1958)) and $M_2=0.20$ cm^{-1}, we find from (33) that terms of order B^6 contribute nearly -12 % for the maximum B^2. Assuming on the average this contribution to be -6 % and the non-Maxwellian correction to be nearly $+20$ %, we get a net correction of nearly $+15$ % to the value of C as given by (17) for constant transport mean free path. Our calculated value of $C\bar{D}_0$ is 2.0 cm^4 sec^{-1}, which is nearly half the experimental value. Probably the experimental value is too high. The value of $\bar{D}_0$ as measured by Iyengar *et al.* in the same experiment is somewhat lower than the value quoted by other workers (Koechlin *et al.* (1956)). Iyengar *et al.* have also measured the variation of the diffusion cooling constant in the temperature range from room temperature to 140° C, and they find that it increases very rapidly with the temperature, at 140° C it has increased by more than a factor of three. This result is hard to understand and reconcile with the theory. In view of all this it would be advisable to repeat these experiments before a satisfactory comparison could be made with the present theory.

(c) *Graphite.*—Since the first measurements of Antonov *et al.* (1956), Beckurts (1957) has made a careful measurement of the diffusion cooling constant. His experimental value of C is 7 ± 1 cm^2. The maximum value of B^2 in his experiments was 5.7×10^{-2} cm^{-2}. Using the value of 2.7 cm. for $\bar{l}_{tr}$ and $M_2 = 0.068$ cm.$^{-1}$ (this value of M_2 is due Nelkin, 1958), we find that the correction due to terms of order B^6 is -7 %. In spite of the fact that here $(B^2)_{\max}$ is small, this correction is comparatively large and this is due to the small value of M_2 for graphite. Taking an average correction of -4 % and a non-Maxwellian correction of $+20$ %, we find that the calculated value of C is 7.0 cm^2, in very good agreement with the experimental value. Contrary to the case of beryllium, this agreement in our view is indeed significant since neither the experimental error is large nor the assumption of a constant transport mean free path in the case of graphite is bad.

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APPENDIX

In the incoherent approximation, the formula for the differential scattering cross section can be written in the form

$$\frac{d^2 \sigma}{d \Omega \, d \omega} = \frac{\sigma_0}{4 \pi} \frac{k}{k_0} e^{-2W} \sum_{n=0}^{\infty} \frac{(2W)^n}{\underline{|n}} G_n(\omega - \omega_0), \tag{A–1}$$

where σ_0 is the free atom cross section, k_0 and k are, respectively, the wave numbers of the incident and scattered neutrons, and $\hbar \omega_0$ and $\hbar \omega$ are the corresponding energies in units of $\hbar \omega_m$, ω_m being the maximum frequency. $2W$ is the Debye-Waller factor and is given by

$$\begin{aligned} 2W &= Q \gamma(0) \\ &= \frac{m}{M} (\omega + \omega_0 - 2 \sqrt{\omega \omega_0} \cos \theta) \gamma(0), \end{aligned} \tag{A–2}$$

where θ is the angle between $\mathbf{k}$ and $\mathbf{k}_0$, M is the mass of the atom and m that of the neutron and $\gamma(0)$ is defined by

$$\gamma(0) = \int_0^1 \coth(\beta\xi) \frac{f(\xi)}{\xi} d\xi, \tag{A-3}$$

$f(\xi)$ is the frequency function and $\beta = \frac{\hbar}{2 k_0 T}$, k_0 here is the Boltzmann's constant.

The notation used here is that of Sjölander (1958). The G-functions are defined in Sjölander's paper (Eqs. II·22 and II·27).

For the moderators under consideration, it is a very good approximation to retain in (A–1) terms up to the order $\left(\frac{m}{M}\right)$. Then (A–1) becomes

$$\frac{d^2\sigma}{d\Omega\, d\omega} = \frac{\sigma_0}{4\pi}\sqrt{\frac{\omega}{\omega_0}} \cdot$$

$$\cdot \left[G_0 + (G_1 - G_0)\frac{m}{M}\gamma(0)(\omega+\omega_0) - \frac{2m}{M}\gamma(0)(G_1 - G_0)\sqrt{\omega\omega_0}\cos\theta\right]. \tag{A-4}$$

From (A–4) it is clear that in the Legendre expansion of $\sum_s(\omega_0 \to \omega,\ \mathbf{\Omega} \to \mathbf{\Omega}')$ only the terms $l=0$ and $l=1$ need be retained. This fact was used in deducing equation (65) from equation (64) in Section 8.

Calculation of M_2.—In the calculation of M_2, we are concerned with only inelastic scattering. In the summation in (A–1), $n=0$ term is, therefore, to be omitted. Performing the angle integration in (A–1) and retaining terms up to $\left(\frac{m}{M}\right)^2$ we have

$$\frac{d\sigma}{d\omega} = \sigma_0 \frac{m}{M}\gamma(0)\left(\frac{\omega}{\omega_0}\right)^{\frac{1}{2}}(\omega+\omega_0)\, G_1(\omega-\omega_0) - \frac{\sigma_0}{3}\left(\frac{m}{M}\right)^2 \gamma^2(0)\left(\frac{\omega}{\omega_0}\right)^{\frac{1}{2}}$$

$$(3\omega^2 + 3\omega_0^2 + 10\,\omega\omega_0)\left\{G_1(\omega-\omega_0) - \tfrac{1}{2}G_2(\omega-\omega_0)\right\}. \tag{A-5}$$

Here, we shall only consider terms of order $\left(\frac{m}{M}\right)$. The expression for M_2 (Paper I, Eq. 4) is

$$M_2 = \frac{1}{(k_0 T)^4}\int_0^\infty dE_0 \int_0^\infty dE\, \Sigma(E_0 \to E)(E_0 - E)^2 E_0\, e^{-E_0/k_0 T}. \tag{A-6}$$

Now

$$\begin{aligned} G_1(\omega-\omega_0) &= \frac{f(|\omega-\omega_0|)}{(\omega-\omega_0)}\frac{1}{\gamma(0)}\cdot\frac{\coth\beta(\omega-\omega_0)-1}{2} && \text{for } |\omega-\omega_0| \leqslant 1 \\ &= 0 && \text{for } |\omega-\omega_0| > 1. \end{aligned} \tag{A-7}$$

In the Debye approximation

$$f(|\omega-\omega_0|)=\frac{3(\omega-\omega_0)^2}{\omega_m^2}. \tag{A–8}$$

Substituting for $\sum_s(E_0\to E)$ from $(A-5)$ (only the first term) and using (A–7) and (A–8) and keeping in mind the units, we have for M_2, the expression

$$M_3=\Sigma_0\frac{m}{M}\frac{3}{(k_0\theta)^3}\frac{1}{(k_0T)^4}\int_0^\infty E_0e^{-E_0/k_0T}\,dE_0\int_0^\infty\left(\frac{E}{E_0}\right)^{\frac{1}{2}}\frac{E+E_0}{E-E_0}\cdot$$

$$\cdot\frac{(E-E_0)^4}{e^{E-E_0/k_0T}-1}\,dE. \tag{A–9}$$

Putting $E-E_0/k_0T=x$ and $E_0/k_0T=\varepsilon$, (A–9) becomes

$$M_2=\Sigma_0\frac{3m}{M}\cdot\left(\frac{T}{\theta}\right)^3\int_0^\infty\varepsilon e^{-\varepsilon}\,d\varepsilon\int_{-\varepsilon,\,-\frac{\theta}{T}}^{\theta/T}x^3(\varepsilon+x)^{\frac{1}{2}}\frac{1}{\varepsilon^{\frac{1}{2}}}(x+2\varepsilon)\frac{1}{e^x-1}\,dx, \tag{A–10}$$

where the lower limit is $-\varepsilon$ if $\varepsilon<\theta/T$, and $-\frac{\theta}{T}$ if $\varepsilon\geqslant\frac{\theta}{T}$.

Eq. (A–10) can also be written as

$$\frac{M_2}{4\,\xi\,\Sigma_0}=\frac{3}{8}\left(\frac{T}{\theta}\right)^3\cdot\int_0^{\theta/T}\varepsilon e^{-\varepsilon}\,d\varepsilon\int_{-\varepsilon}^{\theta/T}x^3(\varepsilon+x)^{\frac{1}{2}}\frac{1}{\varepsilon^{\frac{1}{2}}}(x+2\varepsilon)\frac{1}{e^x-1}\,dx$$

$$+\frac{3}{8}\left(\frac{T}{\theta}\right)^3\cdot\int_{\theta/T}^\infty\varepsilon e^{-\varepsilon}\,d\varepsilon\int_{-\theta/T}^{\theta/T}x^3(\varepsilon+x)^{\frac{1}{2}}\frac{1}{\varepsilon^{\frac{1}{2}}}(x+2\varepsilon)\frac{1}{e^x-1}\,dx, \tag{A–11}$$

where ξ is the mean logarithmic energy decrement and is equal to $\frac{2}{M/m}$. To calculate M_2, $(A-11)$ has to be evaluated numerically. The only property of the solid which enters on the right hand side of (A–11) is the Debye temperature.

We have used here the incoherent approximation but the difference between the value of M_2 in the case of beryllium as calculated by (A–11) and the value obtained by using the one phonon coherent calculation and the two phonon incoherent calculation is very small. The present accuracy of the experiments does not warrant a better calculation of M_2 than that given by (A–11). We might add here a remark for graphite. It is well known that a simple Debye spectrum as given by (A–8) is not at all satisfactory for graphite, and neither is the model of Krumhansl and Brooks very satisfactory as shown by the inelastic scattering cross section calculations for cold neutrons by Kothari and Singwi (1957). Schofield and Hassitt (1958), however, find that if one uses a Gaussian form for the G-function in (A–5), one gets a good agreement with the measured inelastic cross section. In that case M_2 can be calculated as before except that one uses the Gaussian form of the function G, as given by formula II·41 of Sjölander (1958), where the constant Δ_0 can now be adjusted to give the right fit with the experimental values for the cold neutron cross section. We have not yet carried out this programme.

We shall now consider the two limiting cases of (A–11): (i) when the temperature is high, i.e., $T/\theta \rightarrow \infty$ and (ii) when the temperature is low, i.e., $\theta/T \rightarrow \infty$. Case (i) will give us the classical limit for M_2.

Case (i). The main contribution comes from the second term in (A–11). We, therefore, have

$$\frac{M_2}{4\,\xi\,\Sigma_0} \simeq \frac{3}{4}\left(\frac{T}{\theta}\right)^3 \int_0^\infty \varepsilon^2 e^{-\varepsilon}\, d\varepsilon \int_{-\theta/T}^{\theta/T} x^2\, dx, \tag{A–12}$$

since x is small compared to ε and $e^x \simeq 1+x$. The right hand side of (A–12) is equal to unity. Therefore

$$M_2 = 4\,\xi\,\Sigma_0.$$

The above result though rigorously true in the high temperature limit, will probably hold good for $T > \theta$.

Case (ii). $\theta/T \rightarrow \infty$.

In this case it is the first term in (A–11) which gives the main contribution. We, therefore, have

$$\begin{aligned}\frac{M_2}{4\,\xi\,\Sigma_0} &\simeq \frac{3}{8}\left(\frac{T}{\theta}\right)^3 \int_0^\infty \varepsilon^{\frac{1}{2}} e^{-\varepsilon}\, d\varepsilon \int_0^\infty \frac{x^{9/2}}{e^x - 1}\, dx \\ &= \frac{3}{8}\left(\frac{T}{\theta}\right)^3 \cdot \Gamma\left(\frac{3}{2}\right) \cdot \Gamma(11/2)\, \zeta(11/2).\end{aligned} \tag{A–13}$$

It must be remembered that (A–13) gives the correct temperature dependence but as regards the magnitude of M_2, it gives only an order of magnitude answer. The intermediate case when $\frac{\theta}{T} \geqslant 1$, but not very much greater is of practical interest, since for the moderators under consideration this condition is satisfied at room temperature and a few hundred degrees above. In this range it is not possible to derive limiting expressions of the type (A–13) or (A–12).

Variation of $\bar{D}_0$ with temperature.—By definition

$$\bar{D}_0 = \frac{1}{3}\int_0^\infty l_{tr}(E)\, M(E)\, dE \Big/ \int_0^\infty \frac{1}{v}\, M(E)\, dE, \tag{A–14}$$

where

$$l_{tr}(E) = \frac{1}{\Sigma_s(E)\,(1 - \overline{\cos\theta})}, \tag{A–15}$$

and

$$M(E) = \frac{1}{(k_0 T)^2} \int_0^\infty E\, e^{-E/k_0 T}\, dE. \tag{A–16}$$

The high temperature limit, i.e., $T/\theta \gg 1$ is not very interesting. In this case it is easy to show that $\bar{D} \propto T^{\frac{1}{2}}$. We shall consider the other limiting case, i.e., $\theta/T \to \infty$. Since our aim is to know qualitatively the temperature dependence of $\bar{D}_0$, we shall omit $\overline{\cos\theta}$ in (A–15) and also we shall assume that the temperature is so low that only the inelastic scattering contributes. (This will not apply to graphite even for temperatures as low as 100°K.) If we consider only terms up to order $\left(\frac{m}{M}\right)$ in (A–5), then proceeding as before we have

$$\Sigma_s(\varepsilon) = 3\,\Sigma_0 \left(\frac{m}{M}\right) \left(\frac{T}{\theta}\right)^3 \frac{1}{\varepsilon^{\frac{1}{2}}} \int_{-\varepsilon}^{\theta/T} \frac{x}{e^x - 1} (x+\varepsilon)^{\frac{1}{2}} (x + 2\,\varepsilon)\, dx.$$

or

$$\Sigma_s(\varepsilon) \simeq 3\,\Sigma_0 \left(\frac{m}{M}\right) \left(\frac{T}{\theta}\right)^3 \cdot \frac{1}{\varepsilon^{\frac{1}{2}}} \int_0^\infty \frac{x^{5/2}}{e^x - 1}\, dx$$

$$\simeq 3\,\Sigma_0 \left(\frac{m}{M}\right) \left(\frac{T}{\theta}\right)^3 \cdot \frac{1}{\varepsilon^{\frac{1}{2}}} \cdot \Gamma(7/2)\,\zeta(7/2). \tag{A–17}$$

Substituting the above value in (A–14), we have

$$\bar{D}_0 = \frac{1}{3} \left(3\,\Sigma_0 \left(\frac{m}{M}\right) \left(\frac{T}{\theta}\right)^3 \cdot \Gamma(7/2)\,\zeta(7/2)\right)^{-1} \left(\frac{2\,k_0 T}{m}\right)^{\frac{1}{2}} \int_0^\infty \varepsilon^{3/2} e^{-\varepsilon}\, d\varepsilon \Big/ \int_0^\infty \varepsilon^{\frac{1}{2}} e^{-\varepsilon}\, d\varepsilon$$

$$= \frac{1}{3} \left(3\,\Sigma_0 \left(\frac{m}{M}\right) \Gamma(7/2)\,\zeta(7/2)\right)^{-1} \cdot \left(\frac{2\,k_0 T}{m}\right)^{\frac{1}{2}} \left(\frac{\theta}{T}\right)^3. \tag{A–18}$$

Hence

$$\bar{D}_0 \propto \left(\frac{\theta}{T}\right)^3 T^{\frac{1}{2}}. \tag{A–19}$$

Evaluation of γ_{11} *and* γ_{22}.—From equation (9)

$$\gamma_{ik} = \frac{1}{\sqrt{\underline{|i}\;\underline{|i+1}\;\underline{|k}\;\underline{|k+1}}} \int_0^\infty \int_0^\infty dx\, dx' \left\{\Sigma_s(x \to x')\, L_i^{(1)} \cdot (x)\, L_k^{(1)}(x)\, x\, e^{-x} \right.$$

$$\left. - \Sigma_s(x' \to x)\, L_i^{(1)}(x')\, x'\, e^{-x'}\, L_k^{(1)}(x)\right\}.$$

Now $L_1^{(1)}(x) = 2 - x$. The condition of detailed balance is

$$x\, e^{-x} \sum_s (x \to x') = x'\, e^{-x'} \sum_s (x' \to x).$$

Using this condition and the expression for $L_1^{(1)}(x)$ we have for γ_{11},

$$\gamma_{11} = \frac{1}{2} \int_0^\infty \int_0^\infty d\,x\, d\,x' \sum_s (x \to x')\, x\, e^{-x} \left(x^2 - x\,x' - 2\,(x - x')\right)$$

$$= \frac{1}{2} \int_0^\infty \int_0^\infty d\,x\, d\,x' \sum_s (x \to x')\, x\, e^{-x} \left(x^2 - x\,x'\right),$$

since the other integral vanishes. Hence

$$4\,\gamma_{11} = \int_0^\infty \int_0^\infty d\,x\, d\,x' \sum_s (x \to x')\, x\, e^{-x} \,(x - x')^2 \qquad \text{(A–20)}$$

$= M_2$ of paper I, as can be seen by substituting for x and x' in terms of E and E'.

Now $$L_2^{(1)}(x) = x^2 - 6\,x + 6.$$

We have for γ_{22}, the following expression:

$$\gamma_{22} = \frac{1}{12} \int \int d\,x\, d\,x' \left\{ \sum_s (x \to x')\, x\, e^{-x} \left(x^2 - 6\,x + 6\right)^2 \right.$$
$$\left. - \sum_s (x' \to x)\, x'\, e^{-x'} \left(x^2 - 6\,x + 6\right)\left(x'^2 - 6\,x' + 6\right) \right\}.$$

Using the condition of detailed balance and remembering that only those terms of the integrand which are symmetrical in x and x' will give a finite value for the integral, we have

$$\gamma_{22} = \frac{1}{12} \int \int d\,x\, d\,x' \sum_s (x \to x')\, x\, e^{-x} \left[\frac{1}{2} (x - x')^2 (x + x')^2 + 18\,(x - x')^2 \right.$$
$$\left. - 6\,(x - x')^2 (x + x') \right]. \qquad \text{(A–21)}$$

In the $\left(\dfrac{m}{M}\right)$ approximation $\sum_s (x \to x')$ is given by

$$\sum_s (x \to x') = \sum_0 3 \left(\frac{m}{M}\right) \left(\frac{T}{\theta}\right)^3 \cdot \left(\frac{x'}{x}\right)^{\frac{1}{2}} \cdot \frac{x + x'}{x' - x} \left| x' - x \right|^2 \frac{1}{e^{x' - x} - 1}. \qquad \text{(A–22)}$$

In general (A–21) has to be evaluated numerically. Since we are mainly concerned with the value of the ratio γ_{22}/γ_{11}, we shall evaluate (A–21) in the high temperature limit, i.e., $T/\theta \to \infty$, and hope that an exact calculation of γ_{22}/γ_{11} will give a value not very different from that given by (A–23). The proceedure followed here is the same as that in deducing (A–12). Put $x' - x = \varepsilon$, which is small, then

$$\begin{aligned}\gamma_{22} &= \frac{1}{12}\Sigma_0\, 3\left(\frac{m}{M}\right)\left(\frac{T}{\theta}\right)^3 \cdot \int_0^\infty d\,x\, e^{-x}\, 2\,x^2\,(2\,x^2+18-12\,x)\int_{-\theta/T}^{+\theta/T}\varepsilon^2\, d\,\varepsilon \\ &= \Sigma_0\cdot 3\left(\frac{m}{M}\right)\cdot\frac{1}{12}\cdot 2\cdot\left(\frac{\theta}{T}\right)^3\cdot\left(\frac{T}{\theta}\right)^3\frac{1}{3}\,(4\,\Gamma\,(5)+36\,\Gamma\,(3)-24\,\Gamma\,(4)) \\ &= 4\,\Sigma_0\cdot\left(\frac{m}{M}\right) = 2\,\xi\,\Sigma_0 \\ &\qquad = 2\,\gamma_{11}. \qquad\qquad \text{(A–23)}\end{aligned}$$

Similarly one can show that in the high temperature limit $\gamma_{12} = 0$. In general $\gamma_{12} \neq 0$.

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